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Torrefaction of oil palm EFB in the presence of oxygen

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ABSTRACT

Torrefaction of oil palm empty fruit bunches (EFB), waste products from the palm oil industry, was carried out in a fixed-bed tubular reactor in the presence of oxygen at concentrations ranging from 0% to 15%. The effects of oxygen concentration (0%, 3%, 9% and 15%), temperature (493, 523 and 573 K) and biomass size (0.375, 1.5, 3 and 6 mm) on the torrefaction mass and energy yields were investigated. The mass yield decreased with increasing temperature and oxygen concentration, but was unaffected by biomass size. The energy yield decreased with increasing oxygen concentrations, but was still between 85% and 95%. The energy yield showed concave profiles with increasing temperature. Mechanistically, it was found that the 'oxidative torrefaction' process occurred in two successive steps or via two parallel reactions, where; one reaction is ordinary torrefaction, and the other is oxidation. Analysis of the reaction kinetics based on the suggested mechanisms produced an activation energy of torrefaction, that was consistent with previous studies.

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1. Introduction

Lignocellulosic biomass is one of the most abundant biomass resources on the earth and can be used as a feed-stock for preparing fuels and chemicals. Some of these technologies are still under development. Due to its availability in Malaysia, oil palm waste is considered to be the best biomass waste [1]. In 2010, Malaysia was the second largest producer of palm oil, producing 17.8 million tones, or 39% of the total world supply. Indonesia was the world's largest producer of palm oil, producing 22.2 million tons of oil, or 48% of the total world supply [2]. In 2010, productive oil palm plantations in Malaysia covered 4.9 million ha, a 3.3% increase from 2009, when productive oil palm plantations covered 4.7 million ha [3]. Types of biomass produced by the oil palm industry include empty fruit bunches (EFB), mesocarp fiber, kernel shells, fronds and trunks. EFB, mesocarp fiber and kernel shells are used or discarded at palm oil mills, while fronds and trunks are used or discarded at the plantations. The amount of each type of biomass is summarized in Table 1. Since the current primary energy supply in Malaysia is roughly 70 Mtoe (million tons of oil equivalent), the total oil palm biomass energy potential of 17 Mtoe may contribute considerably to decreasing the consumption of fossil fuels (natural gas, coal and oil). In order to use biomass waste for energy efficiently, the following drawbacks, compared to fossil fuels, must be overcome:

(1) Higher energy consumption during collection.
(2) Heterogeneous and uneven composition.
(3) Lower calorific value.
(4) Quality decay by biodegradation.
(5) Difficult to transport.
(6) High ash content.

A number of options exist to reduce these drawbacks. The most common techniques are pelletization, liquefaction and gasification of the biomass. Pelletization involves drying, chipping, grinding and pelletizing lignocellulosic biomass. Pelletization is the cheapest option, but has some disadvantages that include lower heat values and quality deterioration by moisture (pellet disintegration, moss growth or bioorganic decomposition). Recently, treatment of biomass at a low temperature, around 473–573 K, under an inert atmosphere was found to be effective for improving the energy density and shelf life of the biomass. This treatment was called 'torrefaction' and has been widely applied to wood and grass biomass over the past few years [4–16]. Torrefaction studies have largely been conducted on wood and grass biomass, including wood dusts [10–14,16], beech [4,6,7], eucalyptus [5,11], willow [6–9,13], larch [6,7], bamboo [13] and canary grass [8]. Few reports on the torrefaction of agricultural, lignocellulosic waste species, like straw [6–8,15], were found, despite these waste species being among the most promising renewable resources, particularly in Southern Asia [1]. The authors have previously reported on the torrefaction behavior of three types of oil palm waste [17].

Although torrefaction is one of the most promising methods to improve lignocellulosic solid fuels, the procedure requires thermal

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Table 1
Oil palm biomass wastes and their potential.

Site	Waste type	Generation rate		Plantation area or FFB processed		Annual generation	Moisture	CV (LHV, dry base)	Annual energy (dry base)	
						million t-wet/y	wt%	GJ/t	PJ	Mtoe
Palm plantation	Trunk	40	t-dry/ha-replantation/y	0.08	million ha-replantation	11	71	16.4	52	1.2
	Fronds	15				4.1	71	14.4	17	0.4
	Fronds	6	t-dry/ha-plantation/y	4.49	million ha-plantation in 2008	92.9	71	14.4	388	9.2
Palm oil mill	EFB	0.2	t-wet/t-FFB	85.7	million t-FFB processed in 2009	17.1	65	15.8	95	2.3
	Fiber	0.12				10.3	42	18.3	109	2.6
	Shell	0.05				4.3	17	18.5	66	1.6
	POME	0.6				51.4	–	–	28	0.66
				Total					727	17.3

energy and an inert atmosphere. If it was possible to use flue gas from burners, inert gas and a considerable quantity of energy could be saved, making the process more economically viable. No evidence is available that demonstrates whether torrefaction can be effectively carried out in the presence of oxygen. This information is necessary since flue gas contains varying quantities of oxygen.

In this paper, torrefaction of a lignocellulosic waste was carried out in a fixed-bed tubular reactor in the presence of oxygen ranging from 0% to 15%. The effects of oxygen concentration, temperature and biomass size on the torrefaction mass and energy yields were investigated. The lignocellulosic biomass waste used was EFB in Malaysia. The EFB was dried, ground and sieved before torrefaction.

2. Experimental

2.1. Biomass samples

EFB were collected from an oil palm plantation at Bota in Perak, Malaysia in July 2010. The moisture content was 69.8 wt%. After drying at 378 K for 24 h, the EFB were ground using a mechanical grinder. The moisture content of the dried, ground biomass was 6.6 wt%. The ground powders were sieved into four different size fractions: 0.25–0.50, 1.0–2.0, 2.0–4.0 and 4.0–8.0 mm, which were designated as 0.375, 1.5, 3.0 and 6.0 mm in nominal average diameter.

2.2. Torrefaction experiment

Torrefaction of the biomass samples was carried out at ambient pressure using a horizontal tubular reactor made of stainless steel, with an internal diameter of 46 mm. The entire set-up is illustrated in Fig. 1. A prescribed amount of EFB (2 g) was weighed, and placed in an alumina boat that was placed in the center of the reactor. After flushing the reactor with torrefaction gas for 15 min, the temperature of the reactor was increased to the desired level (493, 523 or 573 K), at a constant rate of 10 deg/min, using an electric furnace surrounding the reactor. The temperature range of 493–573 K was chosen because the selective decomposition of hemicelluloses occurs between 473 and 573 K. The minimum temperature of 493 K was selected because the torrefaction rate below 493 K may have been too low. After torrefaction for 30 min, the heater was turned off and the reactor was left to cool to ambient temperature. The torrefied sample was then recovered, weighed and stored in an air-tight vessel until it was characterized. Throughout the procedure, 0.1 L/min of torrefaction gas was passed through the reactor. This flow rate was determined by referring the flow rate in a previous study [4]. The concentration of oxygen in the gas was adjusted to 0%, 3%, 9% or 15%, to investigate the effect of oxygen concentration on torrefaction. The torrefaction gas was prepared by mixing nitrogen and compressed air under ambient pressure. During each torrefaction experiment, the collection of volatile substances generated in the reactor was attempted using an iced trap as shown in Fig. 1. After each experiment, no condensate was collected in the trap. From previous studies, the major liquid products of torrefaction are water and acetic acid [7]. In the present study, the amount of liquid product formed during torrefaction was too little to collect at the temperature applied.

2.3. Measurements

Four samples of different diameters were used in this study, and their masses and the calorific values were measured before and after torrefaction. Calorific values were measured using a bomb calorimeter, model C2000 series manufactured by IKA Werke. The calorific value obtained from the bomb calorimeter was the high heat value (HHV), which included the latent heat of the vapor emitted from the specimen. Using the results of the torrefaction experiments, three parameters were calculated using the following three equations:

$$y_{\mathrm{M}} = \frac{\text{Mass of solid after torrefaction}}{\text{Mass of EFB used}} \tag{1}$$

$$CV\ \text{ratio} = \frac{CV\ \text{of solid after torrefaction}}{CV\ \text{of EFB used}} \tag{2}$$

$$y_{\mathrm{E}} = y_{\mathrm{M}} \times CV\ \text{ratio} \tag{3}$$

where y_{M} is the mass yield, *CV* is the calorific value and y_{E} is the energy yield.

3. Results and discussion

The calorific value of the un-torrefied EFB was 17.43 MJ/kg. Wahid previously reported a calorific value of 19.1 MJ/kg for EFB [18]. Differences between our study and previous studies may be acceptable because the physical properties of biomass depend numerous variables, including soil conditions and the harvesting season [19].

3.1. Effect of EFB particle size on mass yield

The EFB particle size had little effect on the mass yield. Hereafter the effects of temperature and oxygen concentration on the torrefaction results will be discussed. The results reported in later sections are for 0.375 mm EFB, unless otherwise noted in the text.

The mass yield depends on the biomass type. Bridgeman et al. [8] reported that the mass yields of three types of biomass: reed canary grass, wheat straw and willow, which were 61.5%, 55.1% and 72.0% respectively, when torrefied at 563 K for 30 min. The authors attributed the observed differences to the concentration of hemicellulose in the biomass. Biomass with higher hemicellulose concentrations produced lower mass yields. Hemicellulose concentration in the EFB used in the present study were not obtained, but previous reports suggested concentrations of 22 [20], 28.0 [21] and 35.3 wt% [22]. It is possible that the EFB used in this study had low concentrations of hemicellulose.

Since the mass yield did not depend on EFB particle size, heat transfer is not the rate controlling step. This criterion has been reported by Pyle and Zaror [23]. They proposed an index number, called the Pyrolysis number, defined as:

$$\mathrm{Py} = \frac{\alpha}{k\rho C_{\mathrm{P}} r_{\mathrm{P}}} \tag{4}$$

Another index is the Biot number:

$$Bi = \frac{\alpha r_{\mathrm{P}}}{\lambda} \tag{5}$$

where α is the external heat transfer coefficient in W/(m^2 K), k is the reaction rate constant in s^{-1}, ρ is the density in kg/m^3, C_{p} is the heat capacity in J/(kg K), r_{p} is the radius in m, and λ is the thermal conductivity in W/(m K) of the biomass particle.

Using the criterion, Jalan and Srivastava estimated that biomass pyrolysis up to 873 K is controlled by the intrinsic pyrolysis reaction when particle sizes are less than 1 mm [24]. Prins et al. stated that biomass torrefaction is controlled by the intrinsic torrefaction reaction when biomass particle sizes are less than 2 mm, based on the criterion [6]. In the present case, the border line was larger than 8 mm based on experimental results. Since accurate values of the parameters in Eqs. (4) and (5) are not available in this study, it is difficult to state whether this result can be explained by Pyle and Zaror's criterion. If the parameters reported by Prins et al. were applied to our result, a particle size of 8 mm would be within the reaction controlling zone.

3.2. Effects of temperature and oxygen concentration on mass yield

Fig. 2 presents the relationship between mass yield and temperature at oxygen concentrations of 0%, 3%, 9% and 15%. The mass yield decreased with increasing temperature. Previous reports observed a similar trend [8,11]. The mass yield also decreased with increasing oxygen concentration, even though the mass ratio at 3% oxygen (triangle in Fig. 2) was almost the same as at 0% oxygen (circle in Fig. 2). This suggested that oxidation of EFB occurred during torrefaction and was substantial in an atmosphere of more than 3% oxygen. In other words, in the presence of oxygen EFB undergoes two reactions. The first is ordinary torrefaction, or the decomposition of hemicellulose, and the other is oxidation of all the components of the biomass. Through torrefaction, hemicellulose present in lignocellulose largely decomposes. Arias et al. [5] reported that a shoulder peak of TGA corresponding to hemicellulose substantially disappeared after torrefaction.

3.3. Effects of temperature and oxygen concentration on calorific value

Fig. 3 shows the relationship between calorific value and temperature at oxygen concentrations of 0%, 3%, 9% and 15%. The calorific value increased with increasing temperature. The same trend has been reported previously, where wood and grass-type lignocellulosic biomass samples were used. This trend arises because the carbon content is increased through torrefaction [8]. Surprisingly, the calorific value had no dependency on oxygen concentrations ranging from 0% to 15%. The authors cannot find any report in which torrefaction is conducted in the existence of oxygen. This result implies that EFB undergoes torrefaction and oxidation in parallel during this oxidative torrefaction, and that the two reactions do not interact with each other.

3.4. Effects of temperature and oxygen concentration on energy yield

Fig. 4 shows the relationship between the energy yield and temperature at oxygen concentrations of 0%, 3%, 9% and 15%. The energy yield is a key parameter for understanding how much energy was reserved after torrefaction. At 0%, 3% and 9% oxygen concentrations, the energy yield at 493 K was almost the same as

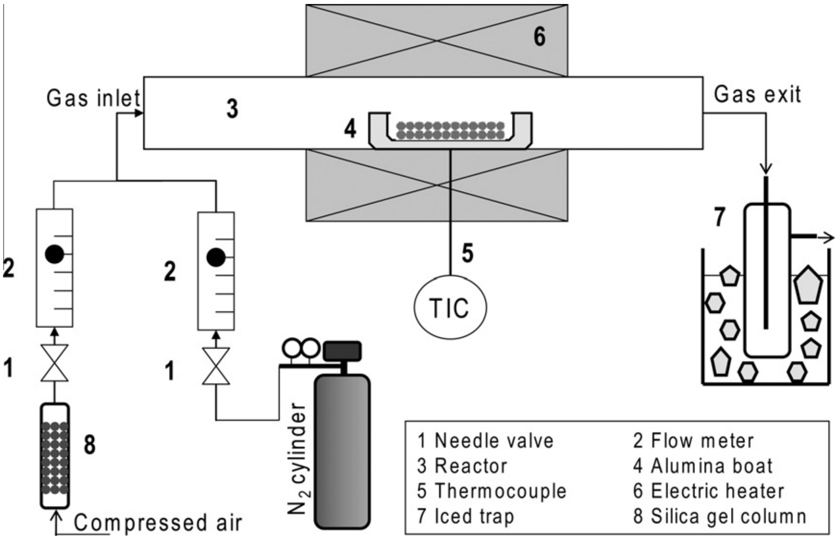

Fig. 1. Experimental apparatus used.

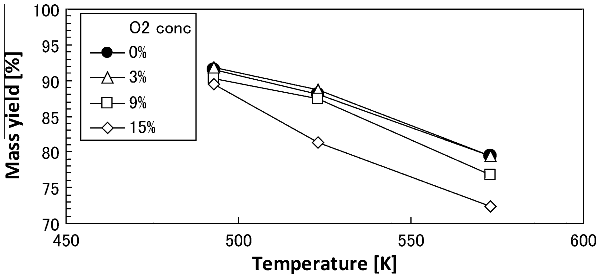

Fig. 2. Effects of temperature and oxygen concentration on the mass yield.

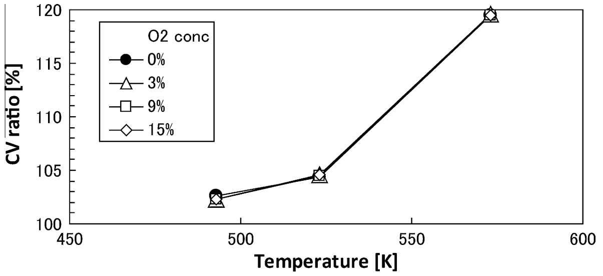

Fig. 3. Effects of temperature and oxygen concentration on the calorific value.

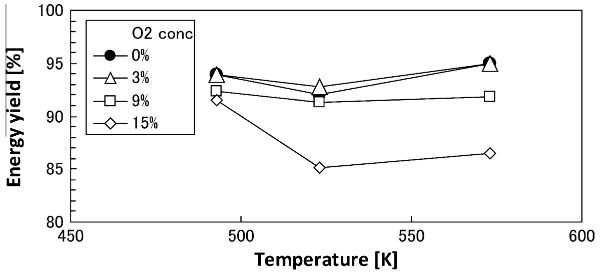

Fig. 4. Effects of temperature and oxygen concentration on the energy yield.

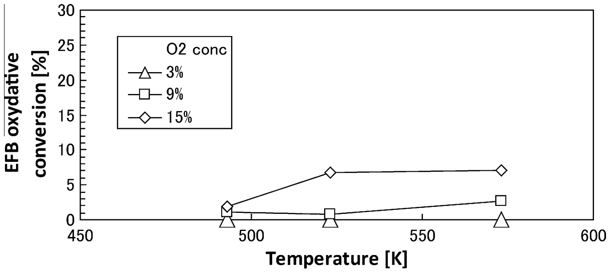

Fig. 5. EFB oxidative conversion.

at 573 K. But, torrefaction at 573 K is preferable because of its larger calorific value than at 493 K. It is worthwhile to mention the temperature dependency of the energy yield. Generally the energy yield decreased with increasing temperature. The trend observed in Fig. 4 is different than previous reports. Further investigation will be required to clarify this matter.

3.5. Reaction kinetics of EFB torrefaction in the presence of oxygen

As discussed in the preceding sections, EFB appears to react in two ways. The first was the decomposition of hemicellulose, or

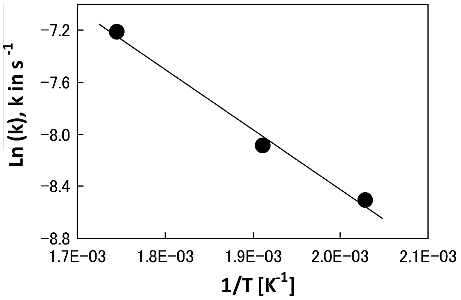

Fig. 6. Arrhenius plot of the torrefaction rate in the absence of oxygen.

ordinary torrefaction, and the other was oxidation of the entire EFB, as follows:

$$-r_{\text{overall}} = (-r_{\text{torref}}) + (-r_{\text{oxy}}) = k_{\text{torref}} C_{\text{HC}}^{l} + k_{\text{oxy}} C_{\text{EFB}}^{m} C_{\text{O}_2}^{n} \tag{6}$$

where $-r$ is the reaction rate of EFB in kg/(m^3 s), k_{torref} is the rate constant of torrefaction in s^{-1}, k_{oxy} is the rate constant of oxidation in m^3/(mol s), C_{HC} is the concentration of hemicellulose in EFB in kg/m^3, C_{EFB} is the concentration of EFB in kg/(m^3 s) and C_{O2} is the concentration of oxygen in mol/m^3.

In order to extract the parameters in Eq. (6), the oxidative portion of the EFB conversion was plotted against temperature in Fig. 5. The oxidative portion plotted in Fig. 5 was calculated by subtracting the total conversion in the absence of oxygen from the total conversion in the presence of oxygen.

The overall torrefaction reaction rate observed in the absence of oxygen can be expressed as follows:

$$-r_{\text{overall}} = -r_{\text{torref}} = -\frac{dC_{\text{HC}}}{dt} = k_{\text{torref}} C_{\text{HC}}^{l} \tag{7}$$

where t is the process time.

According to previous reports on torrefaction kinetics [6,14], the reaction order l is unity. By introducing $l = 1$ and the following expression:

$$C_{\text{HC}} = C_{\text{HC},0}(1 - x_{\text{HC}}) \tag{8}$$

into Eq. (7), Eq. (9) is obtained:

$$\frac{dx_{\text{HC}}}{dt} = k_{\text{torref}}(1 - x_{\text{HC}}) \tag{9}$$

where x is the conversion.

The integrated form of Eq. (9) is:

$$-\ln(1 - x_{\text{HC}}) = k_{\text{torref}} t \tag{10}$$

Introducing the reaction rate data in the absence of oxygen into Eq. (10), the values of k_{torref} were obtained as 2.02×10^{-4}, 3.06×10^{-4} and 7.36×10^{-4} at 493, 523 and 573 K, respectively. The rate constant was plotted against $1/T$ to obtain the activation energy, as shown in Fig. 6. The activation energy was 37.3 kJ/mol. Prins et al. [6] reported the activation energy of willow biomass to be 76.0 kJ/mol. This value was also used and approved by Repellin et al. [14] for simulating the torrefaction behavior of spruce and beech. The differences between our measurements and those reported previously may be acceptable if two factors are taken into account. The first factor is that different types of biomass were used. Their biomass was wood, and our biomass was palm waste. The second factor is that different experimental techniques were applied. Prins et al. used a TG instrument, and we used a tubular plug flow type reactor.

4. Conclusions

Torrefaction of oil palm EFB was carried out in the presence of oxygen to investigate the effects of different torrefaction conditions on the mass and energy yields obtained. The mass yield decreased with increasing temperature and oxygen concentration, but was unaffected by biomass size. The energy yield decreased with increasing oxygen concentration, but all yields were between 85% and 95%. The energy yield showed concave profiles with increasing temperature. In other words, similar energy densities were obtained from the lowest torrefaction temperature, 493 K, and the highest temperature, 573 K. However, the higher the temperature, the higher the energy density. In this sense, the highest temperature may be recommended.

Mechanistically, this 'oxidative torrefaction' process was found to occur in either two successive steps or via two parallel reactions, where the first was ordinary torrefaction and the other was oxidation. Analysis of the reaction kinetics based on this view produced the activation energy of torrefaction, which was consistent with previous studies.

It is worthwhile indicating that torrefaction in the presence of oxygen can be carried out without any significant problems. A maximum 7% of biomass was lost through complete oxidation at 15% oxygen.

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